

# PATENT COOPERATION TREATY

## PCT

### INTERNATIONAL PRELIMINARY REPORT ON PATENTABILITY


(Chapter II of the Patent Cooperation Treaty)

(PCT Article 36 and Rule 70)

REC'D 09 MAY 2005

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Applicant's or agent's file reference B03/2000PC		<b>FOR FURTHER ACTION</b>		See Form PCT/PEA/416
International application No. PCT/EP2004/003263		International filing date (day/month/year) 26.03.2004		Priority date (day/month/year) 26.03.2003
International Patent Classification (IPC) or national classification and IPC C07C5/09				
Applicant BASF AKTIENGESELLSCHAFT				
<p>1. This report is the international preliminary examination report, established by this International Preliminary Examining Authority under Article 35 and transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 6 sheets, including this cover sheet.</p> <p>3. This report is also accompanied by ANNEXES, comprising:</p> <p>a. <input checked="" type="checkbox"/> sent to the applicant and to the International Bureau a total of 2 sheets, as follows:</p> <p><input checked="" type="checkbox"/> sheets of the description, claims and/or drawings which have been amended and are the basis of this report and/or sheets containing rectifications authorized by this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions).</p> <p><input type="checkbox"/> sheets which supersede earlier sheets, but which this Authority considers contain an amendment that goes beyond the disclosure in the international application as filed, as indicated in item 4 of Box No. I and the Supplemental Box.</p> <p>b. <input type="checkbox"/> (sent to the International Bureau only) a total of (indicate type and number of electronic carrier(s)) , containing a sequence listing and/or tables related thereto, in computer readable form only, as indicated in the Supplemental Box Relating to Sequence Listing (see Section 802 of the Administrative Instructions).</p>				
<p>4. This report contains indications relating to the following items:</p> <p><input checked="" type="checkbox"/> Box No. I Basis of the opinion</p> <p><input type="checkbox"/> Box No. II Priority</p> <p><input type="checkbox"/> Box No. III Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</p> <p><input checked="" type="checkbox"/> Box No. IV Lack of unity of invention</p> <p><input checked="" type="checkbox"/> Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</p> <p><input type="checkbox"/> Box No. VI Certain documents cited</p> <p><input type="checkbox"/> Box No. VII Certain defects in the international application</p> <p><input type="checkbox"/> Box No. VIII Certain observations on the international application</p>				
Date of submission of the demand  02.12.2004		Date of completion of this report  06.05.2005		
Name and mailing address of the International preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465		Authorized Officer  Holzwarth, A  Telephone No. +49 89 2399-7269		



**INTERNATIONAL PRELIMINARY REPORT  
ON PATENTABILITY**

International application No.  
PCT/EP2004/003263

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**Box No. I Basis of the report**

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1. With regard to the **language**, this report is based on the international application in the language in which it was filed, unless otherwise indicated under this item.
- ☐ This report is based on translations from the original language into the following language , which is the language of a translation furnished for the purposes of:
- ☐ international search (under Rules 12.3 and 23.1(b))
  - ☐ publication of the international application (under Rule 12.4)
  - ☐ international preliminary examination (under Rules 55.2 and/or 55.3)
2. With regard to the **elements\*** of the international application, this report is based on *(replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report):*

**Description, Pages**

1-19 as originally filed

**Claims, Numbers**

1-11 received on 02.12.2004 with letter of 02.12.2004

**Drawings, Sheets**

1/5-5/5 as originally filed

- ☐ a sequence listing and/or any related table(s) - see Supplemental Box Relating to Sequence Listing
3. ☐ The amendments have resulted in the cancellation of:
- ☐ the description, pages
  - ☐ the claims, Nos.
  - ☐ the drawings, sheets/figs
  - ☐ the sequence listing (*specify*):
  - ☐ any table(s) related to sequence listing (*specify*):
4. ☐ This report has been established as if (some of) the amendments annexed to this report and listed below had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).
- ☐ the description, pages
  - ☐ the claims, Nos.
  - ☐ the drawings, sheets/figs
  - ☐ the sequence listing (*specify*):
  - ☐ any table(s) related to sequence listing (*specify*):

\* If item 4 applies, some or all of these sheets may be marked "superseded."

**INTERNATIONAL PRELIMINARY REPORT  
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**Box No. IV Lack of unity of invention**

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1. ☒ In response to the invitation to restrict or pay additional fees, the applicant has:
- ☐ restricted the claims.
  - ☒ paid additional fees.
  - ☐ paid additional fees under protest.
  - ☐ neither restricted nor paid additional fees.
2. ☐ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.
3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is
- ☐ complied with.
  - ☒ not complied with for the following reasons:  
**see separate sheet**
4. Consequently, this report has been established in respect of the following parts of the international application:
- ☒ all parts.
  - ☐ the parts relating to claims Nos. .

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**Box No. V Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

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1. Statement

Novelty (N)	Yes: Claims	1-11
	No: Claims	
Inventive step (IS)	Yes: Claims	1-11
	No: Claims	
Industrial applicability (IA)	Yes: Claims	1-11
	No: Claims	

2. Citations and explanations (Rule 70.7):

**see separate sheet**

**Re Item IV**

**Lack of unity of invention**

Reference is made to the following document:

D1: KANG J H ET AL: 'Selective Hydrogenation of Acetylene on TiO<sub>2</sub>-Added Pd Catalysts' JOURNAL OF CATALYSIS, ACADEMIC PRESS, DULUTH, MN, US, vol. 208, no. 2, 10 June 2002 (2002-06-10), pages 310-320, XP004465783 ISSN: 0021-9517

**This Authority considers that the application is not unitary, hence does not meet the requirements defined in Rules 13.1 and 13.2 PCT, because of the following reasons:**

Claim 1 contains 3 embodiments, which relate to supported Pd-La-catalysts (embodiment 1), Pd-Ti-K-catalysts (embodiment 2), Pd-La-Si-catalysts (embodiment 3). Embodiments 1 and 3 are linked by the common concept that the catalysts both contain 0.035 - 5.2 wt% lanthanum as an additional component next to Palladium.

The common concept of the other embodiments is the following:

A supported Palladium catalyst consisting of 0.05 - 2.0 wt% Palladium and at least one further component with a concentration of 0.045 - 1.8 wt%.

**This common concept is not novel as**

D1 (abstract, experimental, figure 5) discloses a palladium catalysts consisting of a support (SiO<sub>2</sub>), 1 wt% palladium and 0.45 wt% titanium (molar ratio of Pd:Ti = 1:1). This catalyst is used for the selective hydrogenation of acetylene to ethylene.

The catalysts of D1 solve the same problem as the catalysts in claim 1, which is to improve the selectivity to ethylene in acetylene hydrogenation over supported catalysts which contain just Palladium. Therefore there is also **no corresponding special technical feature**.

**As no corresponding special technical feature and no single general inventive concept link the embodiments 1 - 2, the application does not meet the requirements of unity of invention as defined in Rules 13.1 and 13.2 PCT.**

**This Authority therefore considers that there are 2 inventions covered by the claims**

**indicated as follows:**

**I:** The content of claim 1-11 relating to embodiment 1 and 3 (**Pd-La and Pd-La-Si catalysts**).  
Which is covered by the following claims:  
claim 1 (partly) - claim 3 (partly), claim 5, claim 6 (partly) - claim 7 (partly), claim 9, claim 10 (partly) - claim 11 (partly)

**II:** The content of claim 1-11 relating to embodiment 2 (**Pd-Ti-K catalysts**).  
Which is covered by the following claims:  
claim 1 (partly) - claim 3 (partly), claim 4, claim 6 (partly) - claim 7 (partly), claim 8, claim 10 (partly) - claim 11 (partly)

**Re Item V**

**Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1.1** The document **D1** (see above) is regarded as being the closest prior art.  
The difference of claim 1 to D1 is that titanium is replaced by lanthanum (invention I) and that potassium is added to the Pd-Ti catalyst of D1 (invention II).

**The subject-matter of the claims 1-11 relating to invention I and II is therefore new (Article 33(2) PCT).**

**1.2** The problem to be solved by the inventions I and II may be regarded as providing in the case of invention I alternative acetylene hydrogenation catalysts to the catalysts of D1, in which titanium is replaced by lanthanum. In the case of invention II the performance of Pd-Ti catalysts of D1 is improved by addition of potassium. These solutions can be regarded as not obvious.

**The solution proposed in the claims 1-11 relating to invention I and II is considered as involving an inventive step (Article 33(3) PCT).**

**1.3** The subject matter of the dependant claims 2-11 relating to invention I and II, can equally be considered as novel and inventive and therefore satisfy the requirements of Art. 33(2) and 33(3) PCT.

**INTERNATIONAL PRELIMINARY  
REPORT ON PATENTABILITY  
(SEPARATE SHEET)**

International application No.

**PCT/EP2004/003263**

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## Amended set of claims

1. A palladium catalyst consisting of a support and
- 5 from 0.05 to 2.0% by weight, based on the supported catalyst, of palladium and from 0.035 to 5.2% by weight, based on the supported catalyst, of lanthanum,
- or
- 10 from 0.05 to 2.0% by weight, based on the supported catalyst, of palladium, from 0.02 to 1.0% by weight, based on the supported catalyst, of titanium and from 0.0002 to 7.4% by weight, based on the supported catalyst, of potassium,
- or
- 15 from 0.05 to 2.0% by weight, based on the supported catalyst, of palladium, from 0.035 to 5.2% by weight, based on the supported catalyst, of lanthanum and from 0.0001 to 0.065% by weight, based on the supported catalyst, of silicon.
- 20 2. A process for preparing a palladium catalyst according to claim 1 by impregnating a support in tetra amine palladium hydroxide aqueous solution followed by drying and calcination and impregnating the support with precursor solutions containing precursors of the further metals.
- 25 3. A process according to claim 2, wherein the Pd-La-, Pd-Ti- catalyst is prepared by impregnating the Pd-catalyst in corresponding precursor solution followed by drying and calcination.
- 30 4. A process according to claim 3, wherein a Pd-Ti-K-catalyst is prepared by impregnating the Pd-Ti-catalyst in potassium precursor solution followed by drying and calcination.
- 35 5. A process according to claim 2 or 3, wherein a Pd-La-Si-catalyst is prepared by Si-CVD on a Pd-La-catalyst, pre-reduced at 350 to 700°C, followed by oxidation at room temperature.

6. A process according to claims 2 to 5, wherein the producing of the catalysts includes the reduction process at 300 to 600°C for 1 to 5 hours.
- 5 7. A process according to claims 2 to 6, wherein the La-precursor is lanthanum nitrate hydrate and the titanium precursors are chosen from a group consisting of  $\text{Ti}(\text{O}-i\text{Pr})_2(\text{DPM})_2$ , titanium ethoxide, titanium oxide acetylacetonate and titanium butoxide.
- 10 8. A process according to claim 4, wherein the K-precursor is potassium nitrate.
9. A process according to claim 5, wherein the Si-precursor is chosen from the group consisting of tetrahydrosilane, triethylsilane, tripropylsilane and phenylsilane.
- 15 10. A continuous process for the selective hydrogenation of acetylene to ethylene in the presence of a catalyst according to claim 1, wherein 0.5 to 2.0% by weight of acetylene in ethylene/acetylene gas mixture is used, the reaction temperature is 30 to 120 °C and the flow rate of the gas mixture is 200 to 2500 ml/min×g catalyst.
- 20 11. The use of a palladium catalyst as defined in claim 1 in the selective hydrogenation of acetylene.